Paper

RESUSPENSION STUDIES IN THE MARSHALL ISLANDS

Joseph H. Shinn, Donald N. Homan, and William L. Robison*

Abstract—**The contribution of inhalation exposure to the total dose for residents of the Marshall Islands was monitored at occasions of opportunity on several islands in the Bikini and Enewetak Atolls. To determine the long-term potential for inhalation exposure, and to understand the mechanisms of redistribution and personal exposure, additional investigations were undertaken on Bikini Island under modified and controlled conditions. Experiments were conducted to provide key parameters for the assessment of inhalation exposure from plutonium-contaminated dust aerosols: characterization of the contribution of plutonium in soil-borne aerosols as compared to sea spray and organic aerosols, determination of plutonium resuspension rates as measured by the meteorological flux-gradient method during extreme conditions of a bare-soil vs. a stabilized surface, determination of the approximate individual exposures to resuspended plutonium by traffic, and studies of exposures to individuals in different occupational environments simulated by personal air sampling of workers assigned to a variety of tasks. Enhancement factors (defined as ratios of the plutonium-activity of suspended aerosols relative to the plutonium-activity of the soil) were determined to be less than 1 (typically 0.4 to 0.7) in the undisturbed, vegetated areas, but greater than 1 (as high as 3) for the case studies of disturbed bare soil, roadside travel, and for occupational duties in fields and in and around houses.**

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Key words: Marshall Islands; plutonium; soil; inhalation

INTRODUCTION

A STUDY of inhalation exposure from plutonium-contaminated soil was conducted on Bikini Atoll to provide the parameters for a rigorous assessment of the inhalation dose to humans. The study was needed for improved dose assessments through field observations of total suspended particulates ("mass-loading," $\mu g\ m^{-3}$), aerosol size, and radioactivity per unit mass. In addition, this study undertook to describe the distribution of suspended radioactivity within the respirable size range and how either the concentrations of radioactivity or the particle size distributions vary with surface conditions and local resuspending (dust-lifting) mechanisms.

* Health and Ecological Assessment Division, Lawrence Livermore National Laboratory, Livermore, CA 94551.

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Our investigations conducted on Bikini and Enewetak Atolls in the Marshall Islands provided data that have implications not only for the local dose assessment concerning rehabitation of those sites, but that confirm observations elsewhere regarding inhalation exposure due to plutonium resuspension. This research was supported by U.S. Department of Energy, Office of Health and Environmental Research, and is compiled here from a series of older internal reports.

BACKGROUND

A study conducted on Bikini Island in May 1978, provided a more complete set of data following earlier studies on Enjebi Island of Enewetak Atoll in February 1977 (Homan et al. 1978). The Bikini Island study utilized extensive soil sampling and *in situ* gamma spectroscopy to determine isotope concentrations in soil. Also various air sampling devices were used to determine particle size distribution and mass loading, and micrometeorological techniques were used to determine aerosol fluxes. Subsequent wet chemistry analysis provided radionuclide and elemental concentrations in collected aerosols. Four simultaneous experiments were conducted: (1) a characterization of the normal (background) suspended aerosols and the contributions from sea spray off the windward beach leeward across the island using Na and Mg as tracers for sea spray and an array of air samplers; (2) a study of resuspension of radionuclides from a field purposely laid bare by bulldozers as a worst-case condition followed by detailed air sampling; (3) a study of resuspension of radioactive particles by vehicular and foot traffic using an integrating nephelometer and air sampling along a road; and (4) a study of personal inhalation exposure using small air samplers carried by volunteers during their daily routines. Less complete, unreported studies similar to (1) and (2) had been performed previously on Enjebi Island at Enewetak Atoll and background studies similar to (1) were performed later on Eneu Island at Bikini Atoll.

METHODS

Soil samples were collected for analysis of radionuclide concentrations. ^{238}Pu, $^{239+240}Pu$ and ^{241}Am concentrations were determined by isotope dilution and alpha spectrometry. These analyses were performed by LFE Corporation. Also, because the ratios ^{241}Am:$^{239+240}Pu$ and ^{238}Pu:$^{239+240}Pu$ are constant on Bikini, it was possible to estimate plutonium soil con-

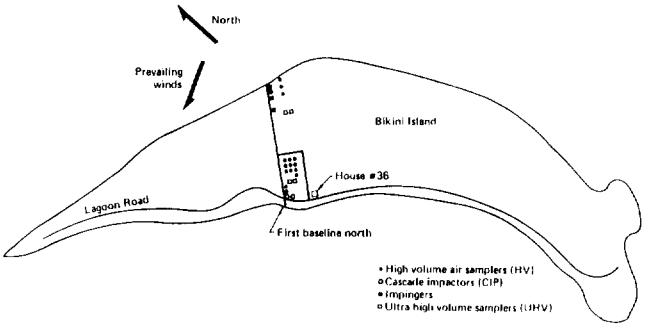

Fig. 1. Plan view of instrument locations on Bikini Island.

centrations by measuring ^{241}Am soil concentrations using a gamma spectroscopy system consisting of a planar, high-purity germanium diode which attained a minimum detectability for ^{241}Am less than 0.037 Bq g^{-1} (Kirby et al. 1977). The detector was mounted facing downward on a tripod so that the volume of soil integrated was contained in a circle of 3 m radius and 5 cm depth. Because the nuclear events causing the original contamination of Bikini Island were far removed, the fallout was relatively evenly dispersed across the Island.

Impingers were used to collect soluble sea spray aerosols in a 250-mL distilled water trap similar to the method of Hsu and Whelan (1976). Air flow rates were 0.36 m^3 h^{-1} (6 L min^{-1}) through the water trap and measured amounts of water were added each day to replace evaporated water (nominally 40 mL d^{-1}). Impingers were set at four tower locations along a 60 m wide clearing from the windward beach inland, spaced at 3, 26, 52, and 102 m from the high-tide waterline and at 1 m and 4 m above the ground. Elemental analysis on the remaining water was obtained by inductively-coupled plasma-optical emission spectroscopy for Na, K, Mg, Ca, and Zn and by a standard autoanalyzer (Technicon) for Cl.

The major particle collection system was an array of 14 standard, high-volume (HV) air samplers, with 8 × 10-inch filters, and two cascade impactors, all using Gelman type AE glass fiber filters. Flow rates for HV (General Metal Works) were monitored at a pressure tap on the fan; discharge was kept at 100 m^3 h^{-1} (60 cfm). The lapsed time of filter operation was recorded for each HV and cascade impactor by counting pulses from a crystal-controlled clock activated by a pressure-sensitive switch. Cascade impactors (CIP) were the 5-stage, jet-plate type.[†] Three HV with the air inlets at 1.1 m above ground were located on a line from the windward beach inland at 5, 70, and 158 m from the high-tide waterline; the latter two were beneath a coconut grove canopy. The tree canopy was expected to shelter the soil surface from wind and to provide a stabilized soil surface. One HV was located downwind of a road at the traffic study site, and ten HV and two cascade impactors were placed in a square array nominally 10-m apart in the middle of a field (1 hectare area) cleared by bulldozing. Special chemistry methods[‡] were employed on the filters to determine the concentration of the stable elements Na, K, Ca, Mg, S, and Cl, and the radioactive isotopes ^{238}Pu, $^{239+240}$Pu, ^{90}Sr, ^{137}Cs and ^{241}Am. Filter blanks were used to correct the stable elements. See map, Fig. 1, for locations of instruments.

In addition to the HV, three non-standard ultra-high volume air samplers (UHV) were used having air inlets at 1.5 m height. Flow-rates, nominally 2,550 m^3 h^{-1} (1,500 cfm), were monitored both by a pitot tube pressure tap on the fan discharge and by a modified anemometer-transducer measuring total discharge in the fan outlet, which was ducted 4 m downwind before discharge. Filters were 1 m^2 area of special fiber-type.[§] One UHV was placed in the coconut grove 370 m from the windward high-tide waterline, and two UHV were operated in the cleared field at the downwind edge of the HV array. The UHV provided the advantage of detection of suspended radioactive aerosols at extremely low levels (e.g., worldwide background) in a matter of a few hours run time. (Locations of UHV are shown on Fig. 1.)

Personal air samplers[‖] were used to determine inhalation exposure rates of individual persons. These personal air samplers (PAs) are small, belt-mounted pumps with a hose connection to a cyclone particle discriminator and filter holder suspended by a chain worn around the neck. Flow rates were 0.11 m^3 h^{-1} (0.064 cfm) and filters were porous-type membrane filters (37 mm diameter, 1 μm pore diameter[¶]). It was found that blank membrane filters inexplicably gained weight with time but that standard deviations within 10% of mass could be achieved where filter blanks from the same lot were monitored for weight gains over the time period of the experiment. The membrane filters were used as a substrate for a scanning electron microscope (SEM) study of particle characteristics. The SEM operated by the LLNL Particle Characterization Facility has a large chamber for specimens (90 mm) and has a resolution of 0.015 μm. In the SEM mount, microprobe chemistry of individual aerosol particles on the membrane filters was accomplished by x-ray fluorescence with a resolution of 160 eV, which provided quantitation of particles containing elements with atomic numbers equal or greater than sodium.

Light-scattering instruments capable of sizing and counting particles within the diameter range 0.3 μm to 10 μm were used in two modes. In the first mode, a particle analyzer[#] was operated so that *in situ* particle-size number-densities could be determined. Particles in air entering the view volume at a rate of 0.43 m^3 h^{-1} (0.25 cfm) are counted individually by the resulting pulses that have a height linear with particle diameter, and classified

[†] Model 65-000, Graseby-Anderson, Atlanta, GA.

[‡] LFE Corporation, Richmond CA.

[§] Microsorban N-98, Aerosol Filter Grade S, Delbag Luftfilter, Holzhauser Strasse 159, 13509 Berlin, Germany.

[‖] Model S, Monitor, Mine Safety Appliances Co., P.O. Box 426, Pittsburgh, PA 15230.

[¶] Corning Costar Corp., 45 Wagag Park, Action, MA 01720.

by means of a conventional 200-channel pulse-height analyzer at a resolution of about 20 channels μm^{-1} (latex sphere calibration). Particle sizes were measured in this mode at the 1.0-m height in the cleared field with the aid of a vane-mounted isokinetic inlet. Particle counts were integrated over 120-s periods at frequent intervals during the experiment. In the second mode, light-scattering instruments were used as continuous monitors of total particle concentration. Two instruments were used in this mode: one, similar to the above but without size discrimination,** was also placed at the 1.0 m height in the cleared field. The second instrument used as a mass concentration monitor was an integrating nephelometer which measures the bulk scattering coefficient. In the 1–10 μm size range, the nephelometer samples ambient air at a flow rate of 17 $m^3 h^{-1}$ (10 cfm) and has a fast response (about 1 s) with a continuous DC analog output that is linearly related to mass concentration using the manufacturer's calibration.†† On Bikini, the nephelometer was used as a dust monitor for foot and vehicular traffic.

Two portable wind systems were utilized. For monitoring wind speed and direction in the open field at the 4.5-m height, a wind speed transmitter with reed switch contact closure each revolution and a wind direction transmitter with 540° dual potentiometer were operated through a battery-powered translator with 30-s filter, and recorded on a dual channel, strip chart recorder with an 8-d spring-driven chart. Wind-speed vertical profiles for aerosol flux estimates were determined using a portable, sensitive, cup-anemometer system‡‡ operated at five height levels up to 4 m for selected 15-min periods (stall speed 0.20 m s^{-1}, distance constant 1.1 m).

RESULTS AND DISCUSSION

Characterization of background aerosols

The contribution of sea spray to aerosol mass loading was investigated thoroughly because it is a diluent of the resuspended mass fraction from the soil, which is the host to plutonium in the atmosphere. The stable element analyses showed that the ratios Cl:Na and Na:Mg were conservative and had values predicted by the composition of seawater (1.8 and 8.3, respectively) (Horne 1969). Other elements, in particular, calcium, were expected to be a tracer for calcareous soil from the parent coral material. The ratio Na:K was somewhat more variable and had a mean value of 11 which is intermediate between those values predicted for sea spray (5.7) and for seawater (27) (Horne 1969; Goldberg et al. 1971). Hence, the concentration of sea spray in the air was calculated by 3.25 × Na concentration and 27 × Mg concentration, based on the ionic composition of seawater (Goldberg et al. 1971). These two values were averaged to minimize random error of determination. The impingers were determined to be 77% efficient for collection of the sea spray by comparison with HV; after this correction and averaging the samples from 1-m and 4-m heights, it was found that the concentration of sea spray was constant over the island to within 52 m from the windward, high-tide waterline (Table 1). The leeward decrease was verified by HV measurements as well, but it occurred in a surprisingly short range compared to sea spray aerosols measured by other investigators (Hsu and Whelan 1976; Yaalon and Lomas 1970; Rossknecht et al. 1973). The rapid drop-off of sea spray from the shoreline was thought to be due to the presence of a massive vegetative barrier along the shore, and we expect that the horizontal flux was already reduced at the shore because the sea spray is mostly generated at the surf line on the coral reef nearly a kilometer upwind from the beach. The HV measurements show that the background sea spray aerosol calculated from Na and Mg concentrations was remarkably uniform throughout the remainder of the island ($\bar{x}$ = 34 $\mu g\ m^{-3}$ S = 8.7, n = 27). The HV results are summarized in Table 2.

Background is here defined as the aerosol concentrations at the 1.1 m height over surfaces that are relatively stabilized and under normal wind conditions. After a week, even the bare soil tended to reach the same average level of dust aerosol concentrations (21 $\mu g\ m^{-3}$) as the coconut grove (Table 2). The background concentration of plutonium in air from resuspension of surface soil was measured in 1972 at Bikini and Eneu Islands (Smith and Moore 1972; Lynch et al. 1972). At that time Bikini and Eneu Islands had been cleared, houses built, and a coconut grove planted on both islands. The vegetation height was very low and the islands were partially barren so that conditions were optimal for maximum resuspension. Smith and Moore reported a plutonium concentration in air at Bikini Island of 3.6 $\mu Bq\ m^{-3}$ (mean of 4 stations) and 1.5 $\mu Bq\ m^{-3}$ (mean of 4 stations). Lynch et al. reported a mean plutonium concentration in air at Bikini of about 1.3 $\mu Bq\ m^{-3}$. All of these results are consistent with our observations listed in Table 2.

An analysis of the personal dosimeter data (discussed later) showed that about 10% of the background dust aerosol was organic. $^{239+240}Pu$ concentration (μBq

Table 1. Variation of Na and Mg aerosols and sea spray with distance from the waterline (impingers, effectively at a 2-m height).

Distance (m)	Na ($\mu g\ m^{-3}$)	Mg ($\mu g\ m^{-3}$)	Calculated sea spray ($\mu g\ m^{-3}$)
3	56.2	6.17	175
26	13.3	1.45	41
52	10.5	1.21	35
102	10.4	1.30	34
Background (HV)	10.5	1.26	34

Model CI-201, Climet Instruments Co. 1320 W. Coulton, Ave., Redlands, CA 92374.

** Model CI-208A, Climet Instruments Co. 1320 W. Coulton, Ave., Redlands, CA 92374.

†† Model 1560, Meteorology Research Inc. Balforte Instrument Co., Wolfe St, Baltimore, MD 21231-3513.

‡‡ Modified, from C.W. Thornthwaite Associates; 1725 Parvin Mill Road, Pittsgrove, NJ 08318.

Table 2. Summary of mass and plutonium aerosol concentrations on Bikini Island.

Date 1978	Distance to windward shore (m)	Ground cover	Sea spray ($\mu g\ m^{-3}$)	Dust aerosol ($\mu g\ m^{-3}$)	Plutonium concentration ($\mu Bq\ m^{-3}$)[a]	Type (number) of instruments	Wind speed direction[b] ($m\ s^{-1}$)	(deg)
5/6–8	780	Bare soil	—	167	270	CIP (2)	4.7	53
5/9–16	780	Bare soil	—	9	9.1	CIP (2)	4.6	43
5/6–8	600–700	Bare soil	34	136	240	HV (3)	4.7	53
5/10–11	600–700	Bare soil	34	23	13	HV (10)	4.1	52
5/12–16	600–700	Bare soil	35	18	7.0	HV (10)	4.6	33
5/8–16	70–160	Coconut trees	34	21	2.4	HV (2)	4.6	45
5/8–16	820	Road	33	41	16	HV (1)	4.6	45
5/8–16	5	Shrubs	40	8	1.1	HV (1)	4.6	45
5/10–11	370	Coconut trees	—	—	1.9	UHV (1)	4.1	52
5/12–16	760	Bare soil	—	—	7.8	UHV (2)	4.6	33
Background[c]	>50	Bare soil	34	21	9.5	CIP (2), HV (20), UHV (2)	4.6	33
Background[c]	>50	Coconut trees	34	21	2.2	HV (2), UHV (1)		

[a] One μBq = 27.027 aCi, and one aCi (attocurie) = 10^{-18} Curie.
[b] Wind measurements recorded for the 4.5 m height at a station in the open field.
[c] Averaged over the surfaces that are stabilized.

m^{-3}) was a factor of 4.3 greater over bare soil than in the coconut grove. (Soil activity was 0.57 and 0.30 Bq g^{-1}, respectively, which is not significantly different within the normal variation encountered.)

The net vertical flux of plutonium is used in risk assessments to estimate the residual remaining on the surface after many years of erosion, especially where downward migration appears to be negligible. If we examine the vertical fluxes of plutonium ($\mu Bq\ m^{-2}\ s^{-1}$) the ratio of fluxes from the two sites will be proportional to the ratio of their wind friction velocities, u', where

$$u' = C_D U_1, \quad (1)$$

and C_D is a drag coefficient equal to 0.106 in the coconut grove and 0.077 in the bare field as determined by our wind profile measurements and U_1 is the wind speed at the 1 m height, which was 4.1 times greater for the bare field than the coconut grove. By eqn (1), the ratio of friction velocities is 3 times greater in the bare field than in the coconut grove. The ratio of their plutonium fluxes is also proportional to the ratio of their concentrations; hence, the plutonium flux is a factor of 4.3 × 3 = 12.9 greater in the stabilized bare field than in the coconut grove. In our previous work, we calculated plutonium aerosol flux with the equation,

$$F = K\,(d\chi/dz) = -pku'\chi, \quad (2)$$

where p is the exponent of a presumed power-law distribution of plutonium with height z (negative sign indicating decreasing concentration with height), k is Karman's constant equal to 0.4, and χ is the mean plutonium concentration in the height range from 0.5 to 2.0 m (Anspaugh et al. 1975). The exponent p is the slope of the plutonium concentration vs. height on a log-log scale. The impinger measurements at the 1-m and 4-m heights along the 60-m wide clearing parallel to the mean wind direction showed a p-value of 0.55 for calcium on Bikini Island, which we presume is the major host of terrestrial plutonium contamination. Previous work indicates p-values between 0.25 and 0.35 for dust aerosols in Western U.S. (Anspaugh et al. 1975).

Using the local p-value of 0.55 and measured values of u' and the background plutonium concentration, χ (2.2 $\mu Bq\ m^{-3}$), as typical for the coconut grove, we obtain a plutonium resuspension flux of 0.057 $\mu Bq\ m^{-2}\ s^{-1}$ (1.8 $Bq\ m^{-2}\ y^{-1}$) which compares to 0.74 $\mu Bq\ m^{-2}\ s^{-1}$ (23 $Bq\ m^{-2}\ y^{-1}$) from the stabilized bare field. Dividing the resuspension flux values by the deposition ($Bq\ m^{-2}$, converting $Bq\ g^{-1}$ using soil density 1500 $kg\ m^{-3}$ and depth 0.05 m) we obtain a fractional rate of resuspension of $2.5 \times 10^{-12}\ s^{-1}$ for the coconut grove and $1.7 \times 10^{-11}\ s^{-1}$ for the stabilized bare field. These resuspension rates correspond to resuspension half-times of 8,800 y and 1,300 y, respectively, and are much shorter than the decay half times for plutonium (24,100 y). By comparison, plutonium resuspension rates from a bare field at H-Area, Savannah River Site, South Carolina, was $4.4 \times 10^{-11}\ s^{-1}$ (halftime 5,000 y), and resuspension rates at Nevada Test Site ranged from $3.9 \times 10^{-11}\ s^{-1}$ (halftime 560 y) for a sandy soil site in Plutonium Valley to $6.0 \times 10^{-13}\ s^{-1}$ (halftime 37,000 y) for Little Feller II nuclear detonation site (Shinn et al. 1989).

Resuspension of radioactive particles from a bare field on Bikini Island

On 6 May 1978, a field was chosen for convenience (adjacent to House No. 36) and bulldozed bare of vegetation without stripping the soil. At the middle of the 100-m × 200-m field, the array of instruments (10 HV, 2 CIP, and 2 UHV) were set up in a regular grid covering about one hectare. The upwind fetch to the nearest instrument was 60 m and lateral borders were 30 m wide. During 6–8 May, three HV and two cascade impactors (CIP) were run during the highest resuspension (disturbed) phase immediately after bulldozing, followed by extensive runs with all instruments during the stable phase, 9–16 May. Wind speeds and direction remained relatively constant (Table 2). Plutonium aerosol concen-

tration (μBq m^{-3}) was greater in the period 6–8 May over the period 9–16 May, by a factor of 25 to 30 as shown by the HV and CIP data of Table 2. Because the disturbed surface was stabilized by light rain at the end of the run on 8 May, the cascade impactor data showed significant differences in the plutonium-activity size distribution as shown on Fig. 2 (ordinate $d\chi/d(\ln D)$ in units Bq g^{-1} of dust aerosol, where D is particle diameter). The plutonium activity curves of Fig. 2 are calculated using a log-normal distribution with the median aerodynamic diameters (MAD) and geometric standard deviations (GSD) obtained by fitting cascade impactor data (Table 3). The aerosol size distributions for plutonium activity determined by CIP and the total mass loading (sea spray plus dust) determined by optical particle analyzer were satisfactorily approximated by a log normal distribution with the given GSD values in Table 3. All other MAD values of Table 3 were determined by cascade impactor.

Two typical cases of number density, $dN/d(\ln D)$, and volume density distributions, $dV/d(\ln D)$, determined by the optical particle analyzer over the stabilized bare soil surface on 9 May and 11 May are shown in Fig. 3. (The data points represent averages across size bins.) It should be noted that the optical particle analyzer sees all liquid and solid aerosols, including both dust and sea spray, so that the total mass obtained by integrating the volume distribution and multiplying by a density factor would not be expected to agree with the dry, residual mass loading measured by HV and CIP.

The relatively good agreement obtained between the different measurement systems indicated in Tables 1, 2, and 3 gives us the confidence to draw conclusions about the significance of resuspension for enhanced inhalation exposure in this worst case example. The MAD decreased from about 2.5 μm to 2 μm and the GSD increased from 2.2 to about 2.9. These changes, if the measurements are significant, would amount to a relatively small change in the lung deposition of the particles.

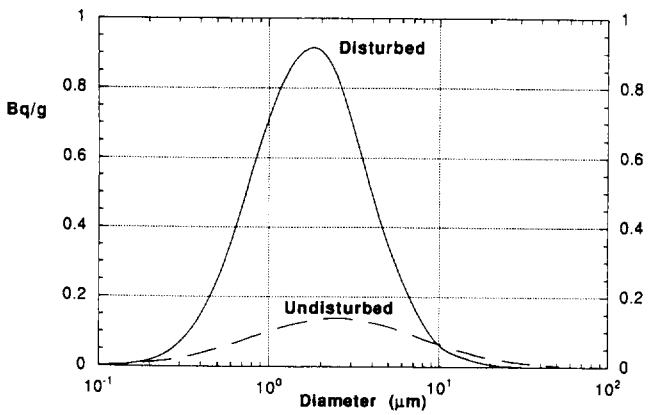

Fig. 2. Plutonium activity size distributions (Bq g^{-1} per Δln D) for disturbed soil (6–8 May) vs. undisturbed soil (stabilized soil 12–16 May).

Table 3. Aerosol size characteristics on Bikini Island determined by cascade impactors and the optical particle analyzer at a height of 1.1 m.

	Disturbed bare soil	Stabilized bare soil
Median aerodynamic diameters (μm)		
Pu activity	1.73	2.46
Pu concentration	2.05	2.43
Mass loading	2.03	2.46
Mass loading—optical	—	2.40 (.11)[a]
Sea spray—Mg	—	2.59
Geometric standard deviation (dimensionless)		
Pu activity	2.16	3.09
Mass loading—optical	—	2.82 (.25)[a]

[a] Optical particle analyzer data with standard deviations in parentheses.

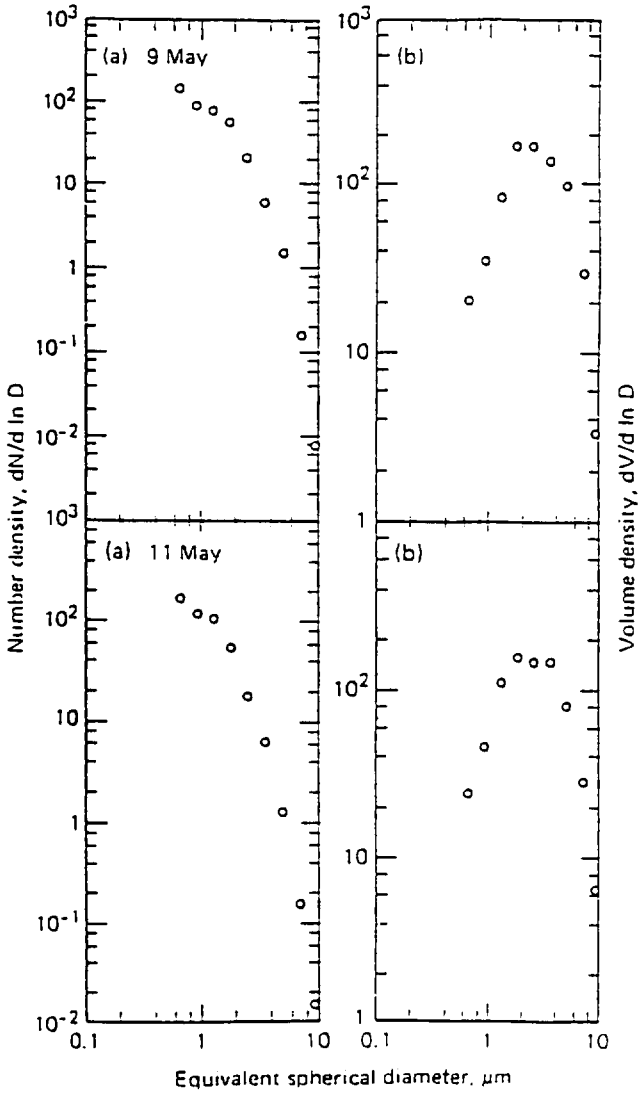

Fig. 3. Typical aerosol size distributions measured by the optical particle counter over the stabilized bare soil surface. Curves (a) are number density (ΔN/ΔlnD; particles cm^{-3}) and curves (b) are volume density (ΔV/ΔlnD; μm cm^{-3}) shown for two different days.

Table 4. Enhancement factors for plutonium activity of aerosols on Bikini Island (HV data).

Site	Date	Soil activity (Bq g^{-1})	Aerosol activity (Bq g^{-1})	Enhancement factor
Disturbed bare soil	5/6–8	0.57	1.8	3.10
Stabilized bare soil	5/10–11	0.57	0.54	0.96
Stabilized bare soil	5/12–16	0.57	0.39	0.69
Coconut grove	5/8–16	0.30	0.12	0.41
Road with traffic	5/8–16	0.15	0.38	2.5

If the total plutonium activity (Bq g^{-1}) is obtained by integration of the curves of Fig. 2, we find that there is, however, a significant change in the aerosol plutonium activity relative to the plutonium activity of the surface soil. Let us define an enhancement factor (EF) as follows:

$$EF = \frac{\text{total aerosol activity (Bq g}^{-1})}{\text{soil activity (Bq g}^{-1})} . \quad (3)$$

Upon investigation of the enhancement factors, we find that the values apparently were less than one under normal conditions (Table 4), probably because of selective resuspension of particles by size. We have found EF values in the range of 0.2 to 1.0 with a median of 0.6 at undisturbed continental sites as well (Shinn 1992). The EF values are expected to be a measure of both the distribution of plutonium with soil particle size and the manner that these particles are aggregated, since EF changes with the degree of disturbance to the soil (Shinn 1992). Organic debris could also dilute the aerosol. The ratio of organic particles to calcareous soil particles remained about constant (10%) as determined by x-ray fluorescence on the PAs filters exposed at this site during the same period. We know from previous studies that one component of organic matter, plant leaves, had a ratio of 10^{-5} plutonium concentration relative to soil and could serve to dilute the inorganic aerosol.

Under dusty conditions, EF values exceed unity, such as in the cases of the disturbed bare soil (3.1) and the road with traffic (2.5). So there are two different factors producing increased plutonium aerosol concentrations (μBq m^{-3}) during unusual resuspension. The aerosol dust concentration increases, but also the plutonium activity increases. For example, averaged over 10 HV instruments, the ratio of plutonium concentration over bare soil on 6–8 May compared to 10–11 May is caused by a 5.91 times increase in dust aerosol concentrations (Table 2) and 3.23 times increase in enhancement factor (Table 4) for a combined effect on aerosol plutonium concentration (239 μBq m^{-3}/12.5 μBq m^{-3}) of 19.1.

Resuspension of radioactive particles by vehicular and foot traffic

The integrating nephelometer was installed with intake at 1.2 m height and 2 m leeward from the position of average tire tracks on a frequently-traveled, one-lane dirt road on Bikini Island. Even though the traditional vehicular traffic of light trucks at low speeds was increased in frequency by our experimental activity, we were interested in characterizing the resuspension of plutonium and inhalation exposure per vehicle pass. The nephelometer provided details on magnitude, duration, and frequency of dust concentrations, while plutonium and dust aerosol concentrations (Table 2), and plutonium activity and enhancement factors (Table 4) were obtained by a co-located HV.

Dust concentrations above background rose in a pulse exceeding 10 s duration where the peak was obtained in a period about 4.5 s after the passage of the vehicle (Fig. 4). This characteristic time to arrival of the peak, regardless of concentration, was determined by X/σ_u where the travel distance X is 2 m and the RMS turbulent velocity σ_u is about one-tenth the local wind speed of 4.5 m s^{-1}. Hence, the dust pulse was traveling by diffusion and not characterized by translation in the wake of the passing vehicle. The dust pulse example of Fig. 4 represents an extreme case (more than 90% of occurrences had lower concentrations) but demonstrates the characteristic peak to mean ratio of 3. 6 and the slow return to background on the trail of the pulse. The amplitude and frequency of dust pulses due to motor vehicle, bicycle, and foot traffic were recorded during 11–15 May. The 68 cases of motor vehicle passes observed showed an approximate log-normal frequency distribution with median peak concentration (above background) of 100 μg m^{-3} and geometric standard deviation of 3.4 (Fig. 5). Bicycle traffic could not be distinguished from foot traffic. In the seven observed cases of foot traffic, we found an approximate median peak concentration above background of 26 μg m^{-3}.

It should be emphasized that the log-normal concentration implies a 5% chance of an exposure to a vehicular-induced peak concentration of 760 μg m^{-3} having a mean concentration 760/3.6 = 211 μg m^{-3} for about 10 s. The plutonium enhancement factor was estimated at 2.5 in this study (Table 4).

Personal inhalation exposure and dosimetry

Until now, the discussion has centered on the (combined) isotope $^{239+240}$Pu, since in fact this is the most important component of inhalation exposure. Extensive soil sampling on Bikini Island has established that a relatively homogeneous mixture of isotopes exists in the soil (Table 5). In the aerosols, some of the isotopes become significantly enhanced (^{137}Cs), but they remain of lesser inhalation-hazard.

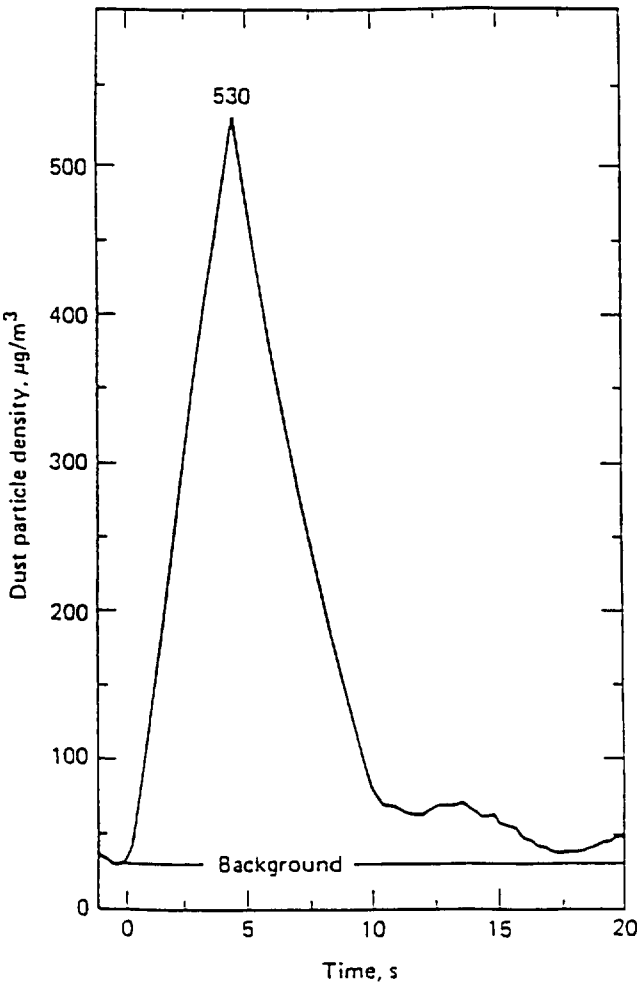

Fig. 4. An example of a dust concentration pulse on the downwind side, 2 m from the edge of a dirt road, showing a peak mass loading of 530 $\mu g\ m^{-3}$ following the passage of a light motor vehicle.

In the case of ^{238}Pu, the concentrations in air were so low that they were probably greatly influenced by the contribution from sea spray.

The *in situ* gamma (ISG) spectroscopy system that measured ^{241}Am in the soil at Bikini was highly correlated to that measured in surface soil samples by special chemistry methods ($R^2 = 0.910$), which gives confidence in both methods. However, data from the ISG system was consistently lower than the soil sampling method by a factor of 0.7 because it integrated a view volume about 5 cm depth and the exponential decrease of isotope concentration with depth gives lower mean values. After correction by a factor 1.44, the ISG method was the primary method for mapping ^{241}Am as a tracer for the source of suspended plutonium isotopes. The horizontal variations in soil isotope concentrations (ISG data) were small enough so that one could justify mean values as local regional values for soil. For example, on the 2-hectare bare field, it was determined that plutonium in

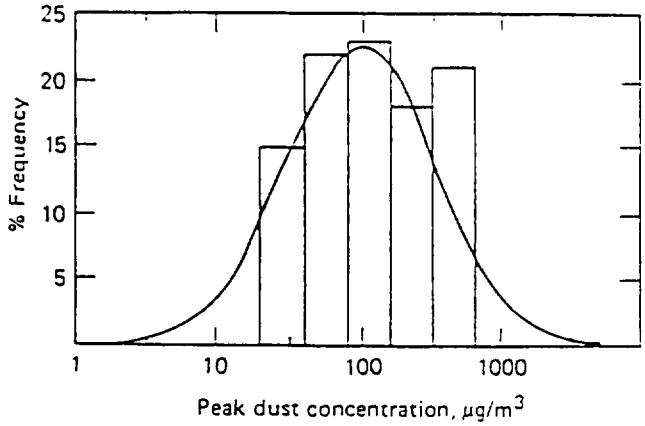

Fig. 5. Frequency of peak dust concentrations on the downwind side of a one-lane dirt road following passages of a light motor vehicle.

surface soil had the mean value 0.57 Bq g^{-1} with an observed range of 0.085 to 1.0, and a fractional standard deviation of 0.57. There was no apparent pattern to the soil concentrations and they exhibit approximately normal, random variation perhaps due to previous tilling and blading locally. (The data did not fit a log-normal distribution any better.)

In the context of living condition effects on exposure, personal air samplers dosimeters (PAs) provided information about inhalation exposure of individuals relative to the reference HV monitors after a necessary adjustment. It was found that the fraction of dust in the total aerosol collected by the PAs was greatest for workers exposed during heavy tilling but was also high for workers exposed in and around houses (Table 6). In this and prior studies, we found that the ratio of PAs Dust:HV Dust has a value of approximately 0.5 where both PAs and HV are sampling the same aerosol cloud of this size particles (2.5 μm MAD) because of the cyclone particle-discriminator on the PAs. Therefore, the enhancement of inhalation exposure by a worker's own actions where the PAs and HV are not sampling the same cloud can be estimated by a personal dosimeter enhancement (PAE):

$$PAE \cong 2 \times (\text{PAs Dust/HV Dust}). \quad (4)$$

Values so computed show significant enhancement (PAE) of inhalation exposure (2.64) during heavy work outdoors by persons sitting or kneeling while digging or using tools on the ground (Table 6). The second highest enhancement (1.86) came from persons with duties in and around the houses. Other work, including heavy tilling, produced inhalation exposures satisfactorily monitored by HV and, thus, their PAE values were close to unity. The main limitation of the PAs data and the derived enhancement values (PAE) is that no information is obtained about the plutonium enhancement factors expressed by eqn (3). It should be recalled that plutonium enhancement factors (EF) of the same magnitude as these PAE were detected by HV (Table 4).

Table 5. Radioactive isotope ratios in soil and aerosols at a stabilized bare field on Bikini Island.

	^{238}Pu[b] / $^{239+240}Pu$	^{241}Am / $^{239+240}Pu$	^{137}Cs / $^{239+240}Pu$	^{90}Sr / $^{239+240}Pu$	$^{239+240}Pu$ (Bq g^{-1})
Soil	0.0013	0.556	9.80	9.15	0.57
FSD[a]	0.56	0.55	0.48	0.52	0.57
Aerosols (HV)	0.050	0.439	61.4	12.8	0.47
FSD[a]	0.64	0.34	0.32	0.51	0.35

[a] Fractional standard deviations ($s/\bar{x}$).
[b] A significant fraction of ^{238}Pu in air was due to sea spray.

Table 6. Comparative effects of living conditions from personal air samplers (PAs) worn by volunteers in various work assignments.

Activity	Outdoors over disturbed bare field	Outdoors over stabilized and vegetated surfaces		Inside and outside around houses
	During tilling	Light work	Heavy work	Light work
Number of volunteers	2	4	3	3
PAs dust fraction[a]	94%	56%	56%	89%
PAs dust (μg m^{-3})	62	12	28	20
PAs dust/HV dust	.46	.55	1.32	.93
PAs enhancement[b]	.92	1.10	2.64	1.86

[a] Personal dosimeter dust is corrected for sea spray but contains about 10% organic matter, both estimated by x-ray fluorescence.
[b] PD enhancement = 2 × (PD dust/HV dust).

Table 7. Inhalation exposure to plutonium ($^{239+240}Pu$) for worst case and best case conditions on Bikini.

Condition	Inhalation rate (m^3 h^{-1})	Dust aerosol (μg m^{-3})	Soil Pu activity (μBq μg^{-1})	Enhancement factor (EF)	Personal enhancement (PAE)	μBq h^{-1} Exposure
Bare field during tilling	1.04	136	0.57	3.10	0.92	230
Stabilized field, heavy work	1.04	21	0.57	0.83	2.64	27
In and around houses, light work	0.83	21	0.57	0.83	1.86	15
Coconut grove, light work	0.83	21	0.30	0.41	1.10	2.4
At roadside, one vehicle/h[a]	0.0023	28	0.15	2.50	(1.0)	0.024

[a] Based on the exposure to one, 10-s, median, vehicular dust-pulse per hour, *not* including background (BG).

Inhalation exposure (μBq h^{-1}) using an inhalation rate (IN-RATE) and previously defined terms may be estimated as follows:

$$\text{INHAL. EXPOSURE} = \text{IN-RATE} \times \text{HV DUST} \times \text{SOIL ACTIVITY} \times \text{EF} \times \text{PAE} \quad (5)$$

$$(\mu\text{Bq h}^{-1}) = (\text{m}^3\,\text{h}^{-1})\,(\mu\text{g m}^{-3})\,(\text{Bq g}^{-1})\,(10^{-6}\,\text{g}\,\mu\text{g}^{-1})\,(10^{6}\,\mu\text{Bq Bq}^{-1}).$$

Using eqn (5), data from Tables 2, 4, and 6, the subtraction of mass loading of sea spray, and the best estimates for the enhancement factors and inhalation rate, we calculated inhalation exposure of $^{239+240}Pu$ for four cases on Bikini (Table 7). Under the worst case condition (during tilling in a disturbed bare field), the inhalation exposure was 230 μBq h^{-1}, and in the best case (light work in a coconut grove), the inhalation exposure was 2.4 μBq h^{-1}. Intermediate values were 27 μBq h^{-1} for heavy work in a bare field, and 15 μBq h^{-1} for light work in and around houses. (In the latter case, we had to use an enhancement factor measured in the nearby field rather than in and around the houses.)

Walking along the road with one vehicular passage per hour produced an estimated 50% chance of additional inhalation exposure of 0.024 μBq h^{-1} (above background) and the soil plutonium activity on the road was notably lower (0.15 Bq g^{-1}) compared to the field (0.57 Bq g^{-1}) (Table 7).

SUMMARY AND CONCLUSIONS

Mass loading (all aerosols) on a HV filter was 55 μg m^{-3} on Bikini Island over stabilized and vegetated surfaces (e.g., in a bare field following rain and in a coconut grove). This compares to 56 μg m^{-3} measured at a vegetated site on Enjebi Island of Enewetak Atoll in February 1977, and a 42 μg m^{-3} weekly average for 10 wk in a coconut grove on Eneu Island of Bikini Atoll May–August 1978. (Wind speeds were comparable, 4–5 m s^{-1}, in all cases.) The more detailed studies at Bikini revealed that 34 μg m^{-3} of the mass loading was salt

Table 8. Plutonium aerosol concentrations on Bikini and Enewetak Atolls compared (winds 4–5 m s $^{-1}$).

Location	Surface description	Plutonium aerosol concentration (μBq m $^{-3}$)	Suspended soil activity (Bq g $^{-1}$)	Surface soil plutonium activity (Bq g $^{-1}$)	Estimated enhancement factor
Normal "Background"					
Bikini	Coconut grove	2.2	0.12	0.30	0.41
Bikini	Stabilized bare soil	9.8	0.47	0.57	0.82
Enjebi	Vegetated field	8.9	0.40[a]	0.90	0.45[a]
Enjebi	Downwind of road	4.0	0.73[a]	1.30	0.56[a]
Unusual Conditions					
Bikini	Field, freshly tilled	2.4	1.8	0.57	3.10
Enjebi	Garden, freshly tilled	2.8	4.0[a]	0.90	4.41[a]
Enjebi	Garden, 1 wk after tilled	1.1	2.3[a]	0.90	2.55[a]
Bikini	Road with traffic	1.6	0.38	0.15	2.50

[a] Calculated by assuming 34 μg m $^{-3}$ sea spray which has been verified by measurement on Bikini.

from sea spray, and that this sea spray contribution remained constant across Bikini Island beyond 20–50 m from the windward beach.

The "background" concentrations of aerosol plutonium on Bikini are comparable to those on Enjebi Island, Enewetak, when one considers the surface soil plutonium activity (Table 8). And, by assuming that Enjebi Island had the same aerosol sea spray background (34 μg^{-3}) as Bikini (which has not been verified by actual measurement) we found that the enhancement factors agree reasonably well. The normal enhancement factor is 0.56, if one assumes that values less than 1 (Table 8) represent normal variations about the mean of 0.56. Apparently, the process of resuspension is preferentially selective by particle size on these atolls to the extent that an aerosol plutonium dilution of 1.8:1 normally occurs.

During unusual surface conditions, such as immediately after tilling, plutonium aerosol activity (normalized by means of the enhancement factor) also agree well. The corresponding enhancement factors were 4.41 on Enjebi Island and 3.10 on Bikini Island.

Plutonium resuspension fluxes due to continuous wind erosion and resuspension were estimated for Bikini by a meteorological flux-gradient equation to be a minimum of 1.8 Bq m^{-2} y^{-1} in the coconut grove and 23 Bq m^{-2} y^{-1} over a bare field stabilized by light rain. Since fields do not remain unvegetated for more than a few months, the coconut grove resuspension flux is probably representative of the island as a whole, even though the wind speeds are one-fourth as high in the coconut grove canopy as in the open.

Particle size distributions measured by both optical and cascade impactor methods show that over the rain-stabilized bare field, the total aerosol size distribution is log-normal with median aerodynamic diameter of 2.4 μm and geometric standard deviation of 3.0, but there is no significant size difference between aerosol plutonium activity and aerosol mass concentration. During the unusual condition of tilling, the size distribution shifts from a median aerodynamic diameter of 2.5 μm to about 2.0 μm with a concurrent increase in plutonium enhancement factor from less than one to 3.1 on Bikini (4.4 on Enjebi). The change in particle size distribution would not contribute to large changes in lung deposition, while the change in plutonium activity would. In the case of a soil disturbed by tilling on Bikini, the plutonium concentration increased by a factor of 19.1 due to a 3.23 $\times$ increase in enhancement factor and a 5.91 $\times$ increase in dust aerosol concentrations.

Vehicular traffic produced dust pulses of nominal 10 s duration in a 4.5 m s^{-1} wind, which were log-normally distributed having time-averaged concentrations above background of 28 μg m^{-3} less than 50% of the time and 211 μg m^{-3} less than 5% of the time. (Peak concentrations were a factor of 3.6 higher.) The plutonium enhancement factor for vehicular traffic was 2.5. Foot and bicycle traffic produced dust pulses about one-fourth as large as vehicular traffic.

Personal air sampling showed that under various exposure conditions, workers inhaled different fractions of inorganic dust and salt, while the organic fraction remained constant at about 10%. Consequently, a personal air enhancement factor was defined to express the effect a worker has by stirring up dust in his own immediate environment.

In conclusion, this study has been as comprehensive as necessary to provide the key parameters for inhalation dose assessment of exposure to plutonium contaminated aerosols. Preliminary dose assessments have been verified now by aerosol measurement methods and at different locations. There remain several unexplained and untested results. It is not yet clear why the aerosol dust concentration is apparently uniform for different surface cover and wind conditions (e.g., coconut grove vs. bare field). It is also not known why the plutonium enhancement factor is less than unity in the normal case, while at the same time, the aerosol plutonium activity and the aerosol mass size distributions are not significantly different. Long-term monitoring on these remote atolls is not yet very practical.

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